



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification 4 :  C08F 26/10, 24/00		A1	(11) International Publication Number: WO 89/05829  (43) International Publication Date: 29 June 1989 (29.06.89)
(21) International Application Number: PCT/US88/04588  (22) International Filing Date: 21 December 1988 (21.12.88)		(74) Agent: GRINNELL, John, P.; Christie, Parker & Hale, P.O. Box 7068, Pasadena, CA 91109-7068 (US).	
(31) Priority Application Number: 138,722  (32) Priority Date: 23 December 1987 (23.12.87)  (33) Priority Country: US		(81) Designated States: AT (European patent), AU, BE (European patent), CH (European patent), DE (European patent), FR (European patent), GB (European patent), IT (European patent), JP, KR, LU (European patent), NL (European patent), SE (European patent).	
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(54) Title: HIGH PERFORMANCE PRESSURE-SENSITIVE ADHESIVE POLYMERS

## (57) Abstract

Pressure-sensitive copolymers based on acrylic monomers are provided with high adhesive performance characteristics by the inclusion of a synergistic amount of an N-vinyl lactam monomer and a glycidyl monomer with the bulk of the monomers being an alkyl acrylate and/or methacrylate esters.

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10 HIGH PERFORMANCE PRESSURE-SENSITIVE  
ADHESIVE POLYMERS

## BACKGROUND OF THE INVENTION

15 The present invention is directed to pressure-sensitive adhesive compositions that exhibit high adhesion to high energy surfaces such as aluminum and possess high tack and outstanding holding power at elevated temperatures. Two key monomers are used in combination in the pressure-sensitive adhesives of this invention to impart unique properties to the resulting polymers. The monomers are at least one glycidyl monomer in combination with at least one N-vinyl lactam.

The use of glycidyl monomers in pressure-sensitive adhesive has been disclosed in the art.

25 U.S. Patent 3,284,423 discloses creep-resistant  
pressure-sensitive adhesive compositions comprising 35-  
75% by weight alkyl acrylate esters containing 6-15  
carbon atoms, 10-60% lower alkyl acrylate, 0.1-10% by  
weight of an ethylenically unsaturated carboxylic acid  
30 and 0.1-10% by weight glycidyl ester.

U.S. Patent 3,893,982 discloses an interpolymer comprising 0.1-15% parts of an ethylenically unsaturated carboxylic acid, 0.1-2% parts of a glycidyl monomer, 35-84.9% parts of an alkyl acrylate or methacrylate and optionally a monomer selected from the group consisting

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1 of alpha-olefins containing 2-10 carbon atoms, vinyl  
esters of alkanoic acids containing 3-10 carbon atoms,  
ethyl and methyl esters of acrylic and methacrylic  
acids, acrylonitrile, methacrylonitrile, styrene and  
5 vinyl chloride where the polymer has a weight average  
molecular weight in the range of 10,000 - 500,000 and  
between 0.01 and 1 parts by weight per 100 parts of the  
copolymer of 1,3-bis(dimethylamino)-2-hydroxypropane to  
cause the cure of the epoxy group.

10 The art has also taught the use of N-vinyl lactams  
in polymers.

15 U.S. Patent 3,728,148 discloses a pressure-  
sensitive adhesive for electrical insulating  
applications comprising of a copolymer of 65-90% by  
weight of an alkyl acrylate ester, 10-30 by weight of a  
N-vinyl lactam and 0-20% by weight of a modifying  
monomer which is copolymerizable with the above. Acidic  
monomers and amides are excluded as they are claimed to  
cause undesirable corrosion.

20 U.S. Patent 4,181,752 discloses an interpolymer  
containing 87% by weight isoctyl acrylate, 8% by weight  
vinyl pyrrolidone, 3% by weight acrylic acid and 2% by  
weight acrylamide.

25 U.S. Patent 4,364,972 discloses a pressure-  
sensitive adhesive tape made by copolymerizing an alkyl  
acrylate ester with 15 to 50 parts by weight of vinyl  
pyrrolidone and having a K-value greater than 100 and  
when crosslinked has a gel-swell in ethyl acetate in  
excess of 600%. Advantages claimed are good adhesion to  
30 automotive paints, rubber and plastic foam layers.

U.S. Patent 4,310,509 discloses a 90/10 2-ethyl  
hexyl acrylate/vinyl pyrrolidone copolymer for making a  
pressure sensitive adherent for complexing with iodine  
for anti-microbial activity.

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1 European Patent Application 130080 discloses an  
emulsion polymerized pressure-sensitive adhesive  
comprising 2-20% by weight N-vinyl lactam and an alkyl  
acrylate ester. The claimed use is good adhesion to  
5 skin under hot and humid conditions.

10 U.S. Patent 4,370,380 is directed to a blend of two  
polymers. One is a copolymer of 88-99% by weight of an  
alkyl acrylate ester with 1-12% by weight of a  
carboxylic acid with a glass transition temperature (Tg)  
15 of less than 0°C. The second polymer is either a homo  
or a copolymer of N-vinyl lactam with a Tg of 20-150°C.  
The blend ratio is 70-99% by weight of the first polymer  
with 1-30% by weight of the second polymer. The  
resultant pressure-sensitive adhesive is disclosed to  
have moisture permeability.

20 U.S. Patent 4,150,197 discloses a water vapor  
permeable pressure-sensitive adhesive comprising a  
copolymer of 79-89% of butyl acrylate, 10-20% by weight  
of N-vinyl lactam and 1-5% by weight of an acidic  
comonomer.

25 None of the patents or applications discloses a  
copolymer containing both a glycidyl monomer and N-  
vinyl lactam monomer.

25 SUMMARY OF THE INVENTION

30 It has now been found that, as part of an acrylic  
and/or methacrylic ester based polymer system, a  
glycidyl monomer and a N-vinyl lactam monomer  
synergistically act to provide unusually high adhesion  
35 to high energy surfaces such as aluminum and stainless  
steel. The pressure-sensitive adhesives of the  
invention are formed of copolymers containing  
essentially no cross-linking when polymerized and which  
contain on a copolymerized basis from about 0.1 to about  
2% by weight of glycidyl monomer, about 1% to about 20%

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1 by weight, preferably from about 1 to about 10% by  
weight of an N-vinyl lactam monomer, from 0 to about 15%  
by weight of an ethylenically unsaturated carboxylic  
acid, from about 55 to about 85% by weight an alkyl  
5 acrylate or methacrylate ester containing from 4 to  
about 12 carbon atoms in the alkyl group, from 0 to 35%  
by weight of an alkyl acrylate or methacrylate ester  
containing less than 4 carbon atoms in the alkyl group,  
and optionally, one or more other monomers employed to  
10 tailor polymer properties, such as glass transition  
temperature, to end use applications.

Such monomers include polystyryl ethyl  
methacrylate, acetoacetoxy ethyl methacrylate, styrene,  
alpha olefins, and vinyl esters of alkanoic acids  
15 containing greater than 3 carbon atoms and mixtures  
thereof. Modifying monomer content can range from 0 to  
about 35% by weight of the total monomers.

The ratio of monomers is selected to provide a  
copolymer with glass transition temperature of less than  
20 about -15°C and a weight average molecular weight of at  
least about 200,000, preferably from about 200,000 to  
about 500,000 as determined by size exclusion  
chromatography using polystyrene for calibration.  
Polymers of the instant invention may be synthesized by  
25 solution, emulsion and bulk polymerization. It is  
presently preferred that they be formed by solution  
polymerization. Polymers are cross-linked to the  
desired extent, prior to use, using heat, ionic  
additives, actinic or electron beam radiation and the  
30 like.

The novel polymers on cross-linking exhibit  
excellent adhesion to high energy surfaces such as  
aluminum as evidenced by increased peel adhesion with  
dwell and superior rivet performance as evidenced by  
35 reduced tenting as described herein.

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1        THE DRAWINGS:

5        FIG. 1 graphically compares 180° peel adhesion on alodine aluminum panels of two adhesives, one which contains both a glycidyl monomer and a lactam monomer to one containing only a glycidyl monomer.

FIG. 2 compares 180° peel on stainless steel as a function of dwell at two temperature conditions, room temperature and 120°C for compositions of the invention to controls.

10        The room temperature dwell was for 20 minutes and 120°C dwell was for 30 minutes. After dwelling for 30 minutes at 120°C, the adhesive on the substrate was allowed to equilibrate to room temperature and 180° peel determined.

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1        DETAILED DESCRIPTION

5        Pressure-sensitive adhesive polymers of the instant invention are prepared by copolymerizing a mixture of monomers comprising from about 55 to about 85% by weight of an alkyl acrylate and/or methacrylate ester containing 4 to about 12 carbon atoms in the alkyl group; from about 0.01 to about 2% by weight of a glycidyl monomer; from about 1 to about 20% by weight, preferably from 1 to about 10% by weight of an N-vinyl lactam monomer; from 0 to 15% by weight, preferably from about 5 to about 13% by weight of an unsaturated carboxylic acid; from 0 to about 35% by weight of an alkyl acrylate and/or methacrylate ester containing less than 4 carbon atoms in the alkyl group and optionally 10      from about 0 to 33% by weight of one or more other comonomers to provide a balance of desirable polymer properties such as glass transition temperature. The precise ratio of the monomers is selected to give a polymer whose glass transition temperature is lower than 15      about -15°C. The polymers of the instant invention have 20      a weight average molecular weight of at least about 200,000, preferably from about 200,000 to about 500,000 as determined by size exclusion chromatography using 25      polystyrene as the calibrator.

25      The alkyl acrylate and methacrylate esters containing 4 to about 12 carbon atoms in the alkyl group useful in forming the polymers of the instant invention include without limitation 2-ethyl hexyl acrylate, isooctyl acrylate, butyl acrylate, sec-butyl acrylate, 30      methyl butyl acrylate, 4-methyl-2-pentyl acrylate, isodecyl methacrylate and the like and mixtures thereof. Isooctyl acrylate and 2-ethyl hexyl acrylate are presently preferred.

35      The glycidyl monomers are glycidyl acrylate, glycidyl methacrylate, allyl glycidyl ether and mixtures

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1 thereof. The presently preferred glycidyl monomer is  
5 glycidyl methacrylate.

5 The N-vinyl lactams monomers which may be used include N-vinyl pyrrolidone, N-vinyl caprolactam, 1-vinyl-2-piperidone, 1-vinyl-5-methyl-2-pyrrolidone, and the like, N-vinyl pyrrolidone is presently preferred.

10 Ethylenically unsaturated carboxylic acids include acrylic acid, methacrylic acid, fumaric acid, and the like.

15 Alkyl acrylate and methacrylate esters containing less than 4 carbon atoms in the alkyl group include methyl acrylate, ethyl acrylate, methyl methacrylate and the like. Methyl acrylate is presently preferred.

20 Other monomers which can be included are polystyryl ethyl methacrylate, acetoacetoxy ethyl methacrylate, alpha olefins such as ethylene and propylene and vinyl esters of alkanoic acids containing more than three carbon atoms as well as mixtures thereof. Such monomer concentrations are in the range from 0 to about 35 percent by weight of the total monomers.

25 The copolymers may be synthesized using solution emulsion, and batch polymerization techniques. It is presently preferred to prepare the copolymers in solution using a mixture of solvents. The present preferred solution polymerization involves the use of blends of ethyl acetate and hexane or ethyl acetate and acetone. The ratio of solvents are adjusted to provide a reflux temperature of from about 68°C to about 78°C. Solids content during polymerization may typically range from about 40% to about 60% in order to achieve the desired weight average molecular weight, and yet achieve viscosities that are manageable in the reactor. Reaction occurs in the presence of free-radical initiators, preferably of the azo type, for example, 30 2,2'-azobis (isobutyronitrile). The polymers formed are 35

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1 solvent soluble polymers with essentially no cross-linking. To this end, the glycidyl monomer is  
5 preferably limited to 2% by weight of the total monomers to avoid the possibility of cross-linking, by opening of the oxirane group, during polymerization or during aging. Polymers can, as desired, be post-polymerization cross-linked using heat, actinic or electron beam radiation and the like.

10 The unique characteristics of the cross-linked pressure-sensitive adhesive copolymers of the instant invention is a dramatic adhesion to high energy surfaces, such to aluminum and stainless steel, as seen by increased peel adhesion values with dwell times and superior rivet performance, as reflected by reduced  
15 tenting. Superior performance requires the presence of both glycidyl and lactam monomers. In the absence of one, poor adhesion to aluminum has been observed, as manifested by poor tenting performance (see Table I). These results are unexpected as a primary purpose in  
20 incorporating the N-vinyl lactam is to increase adhesion to vinyl films and painted surfaces. The purpose of the glycidyl group is normally to introduce a latent functional group which could undergo cross-linking at elevated temperatures under use conditions. This was  
25 realized by the dramatic improvement in elevated temperature performance even as high as 200°C and high levels of shear adhesion failure temperature (SAFT) of polymers containing glycidyl methacrylate as established by Table II. Improved peel adhesion was totally  
30 unexpected. While not bound by theory, it is presently believed that chemical reaction occurs between the epoxy groups and functional groups such as hydroxyl groups on the substrate or a complexation reaction occurs between the epoxy groups and the nitrogen of the lactam, with

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1        ionic groups such as Al+++ on the substrate leading to  
increased bond strength.

5        An application of the adhesives of the instant  
invention is the marking of truck panels and the like  
with an adhesive coated vinyl film. The film is applied  
over a panel which may be a painted or unpainted  
aluminum panel fixed with aluminum rivets. The ability  
of the adhesive to conform to the contour of the rivet  
and not lift appreciably after application is highly  
desirable. A tendency to lift away is known as  
10      "tenting" and the greater the separation from the rivet  
the more unsatisfactory is the adhesive.

15      The SAFT test is a test where the adhesive is  
applied to 0.5" x 1" overlap on stainless steel to which  
a 4.5 lb. roll force applied. After dwell of 24 hours,  
this is placed in an oven and a kilogram load is applied  
under shear conditions and temperature raised from 40°C  
to 200°C at the rate of 1°C per minute. The failure  
temperature is recorded as the shear adhesion failure  
temperature. This is a measure of the cohesive strength  
20      of the adhesive or the ability of the adhesive to  
maintain a bond at elevated temperatures.

25      While not limiting, the following illustrate the  
invention.

25

Example 1

30      A monomer mixture was made up by mixing 423 g. of  
2-ethyl hexyl acrylate, 145 g of methyl acrylate, 3.15  
g. of glycidyl methacrylate, 12.6 g of N-vinyl  
pyrrolidone and 44.1 g of acrylic acid. 157 g. of this  
mixture was introduced to a 2 liter reactor equipped  
with a pitched turbine agitator, a reflux condensor and  
a thermistor. Also added were 73.5 g. of ethyl acetate  
35      and 78.76 g of hexane. The contents of the reactor were

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1        heated to reflux and 0.238 g of Vazo 64, manufactured  
and sold by duPont in 5.0 g of ethyl acetate was added.  
5        Vigorous reflux started in a short time and the contents  
of the reactor were held for 23 minutes. At this time,  
the remaining monomers were mixed with 537.2 g. of ethyl  
acetate, 75.2 g. of hexane and 0.707 g. of Vazo 64 and  
added as a single feed mixture over 3.5 hrs. All  
through the feed, temperature was maintained to keep  
reactor contents under reflux. One hour after end of  
10      feed, 0.17 g. Vazo 64 was added in 5 g. ethyl acetate  
and held for an additional hour. The percentage of  
solids content at the end of reaction was 46.4% and the  
viscosity was 23 Pa.s using #4 @ 12 on a Brookfield  
viscometer.

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Example 2

20        A monomer mixture was made up by mixing 453.6 g. of  
Isooctyl acrylate, 100.8 g of methyl acrylate, 6.3 g. of  
glycidyl methacrylate, 25.2 g of N-vinyl caprolactam,  
44.1 g of acrylic acid and 0.945 g of Vazo 64. 157.5 g  
of this mixture was introduced to the reactor with 78.76  
g of hexane, 78.76 g of ethyl acetate and heated to  
reflux. Once vigorous reflux initiated, the contents  
25      were held for about 12 minutes and the remaining  
monomers added along with 537.24 g of ethyl acetate and  
75.24 g of hexane as a single feed over 3 hours. Two  
hours after the end of feed, the contents were cooled.

30

Example 3 - Tenting Test

35        There was formed as control 1 a polymer containing  
65.7% by weight 2-ethyl hexyl acrylate, 27.3% by weight  
methyl acrylate and 7% by weight acrylic acid. As  
Control 2, there was formed a polymer containing 67% by

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1 weight 2-ethyl hexyl acrylate, 24% by weight methyl acrylate, 7% acrylic acid, and 2% by weight N-vinyl pyrrolidone. As Control 3, there was formed a polymer containing 65.7% by weight 2-ethyl hexyl acrylate, 27.2%  
5 by weight methyl acrylate, 7% by weight acrylic acid and 0.1% by weight glycidyl methacrylate. As Control 4, there was used a polymer containing 65% by weight 2-ethyl hexyl acrylate, 27% by weight methyl acrylate, 7% by weight acrylic acid and 1% by weight glycidyl  
10 methacrylate. These were compared for tenting in a rivet test to the polymers of Examples 1 and 2. As a cross-linker in each instance, there was added 0.2 parts by weight of the polymer of aluminum acetyl acetone. For the rivet tenting test adhesive was transfer coated  
15 from a release liner to a cast vinyl facestock, at a coat weight of 30 g/m<sup>2</sup>. The results are given in Table 1, wherein the lower the value reported, the less tenting, i.e., lifting away from the rivet, occurred.

20 Table 2 compares the polymer of Example 1 electron-beam (EB) cured at a dosage of 30 kiloGray (kGy) and the polymer of Example 2 also EB cured at a dosage of 30 kGy. Examples 1 and 2 exhibited the best combinations of shear and tack.

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TABLE 1

	<u>Control/ Example*</u>	<u>Tenting in Rivet Test, mm</u>
5	Control 1	1.42
	Control 2	1.32
	Control 3	1.37
	Control 4	1.25
10	Example 1	0.58
	Example 2	1.1

\* All the polymers were cross-linked with 0.2 parts per hundred parts of the polymer of Aluminum acetyl acetonate.

15

TABLE 2

		<u>Example 1</u>	<u>Example 2</u>
	<u>180° Peel, NM</u>	<u>EB, 30 kGy</u>	<u>EB, 30 kGy</u>
20	Stainless Steel, 20' Dwell	572	632
	Hard PVC	692	780
<u>Static Shear, min.</u>			
25	20°C/1kg	10000+	9577
	150°C/1kg	5200+	5200+
	200°C/1kg	3300+	3300+
<u>Loop Tack, N/M</u>			
30	600	270	
	<u>SAFT, °C</u>	200°+	200°+

RT Shear, 0.5 x 0.5 inch overlap, Al Facestock  
 SAFT and ET Shear, 0.5 x 1.0 inch overlap, Al Facestock  
 For SAFT, 24 hr. dwell, rate of heating 1°C/minute

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1                   Control 5 to 8 and Examples 4 and 5

5                   There was compared to the product of Example 2 a copolymer containing 67 parts by weight isooctyl acrylate, 25 parts by weight methyl acrylate, 1 part by weight glycidyl methacrylate, and 7 parts by weight acrylic acid (Control 5). Control 5 and Example 2 10 copolymers were coated to a level of 50 g/m<sup>2</sup> onto mylar and cured at an electron beam level of 30 kGy. A comparison of 180° peel on alodine aluminum substrate in 15 Newtons/meter (N/M) as a function of dwell time is shown in Figure 1 and establishes that the combination of glycidyl methacrylate and vinyl caprolactam produce unusually high 180° peels, especially after extended dwell times.

20                  Figure 2 compares 180° peel on stainless steel substrate as a function of dwell at room temperature for 20 minutes and dwell at 120°C for 30 minutes followed by 25 peel testing at room temperature. All polymers employed contained 2-ethyl hexyl acrylate, methyl acrylate and acrylic acid. Control 6 contained no glycidyl methacrylate or N-vinyl lactam. Control 7 contained 0.1 part glycidyl methacrylate but no N-vinyl lactam. Control 8 contained 1 part by weight glycidyl methacrylate but no N-vinyl lactam. Example 4 contained 0.1 part by weight glycidyl methacrylate and 2 parts by weight N-vinyl pyrrolidone. Example 5 contained 0.1 part by weight glycidyl methacrylate and 4 parts by weight N-vinyl caprolactam per hundred parts total monomer. In each instance, the polymers were EB cured 30 at a dosage of 30 kGy. The glycidyl methacrylate in combination and N-vinyl lactam gives better performance 35 in terms of adhesion to high energy surfaces than the individual constituents of the combination.

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1 WHAT IS CLAIMED IS:

1. A pressure-sensitive adhesive comprising a copolymer comprising on a copolymerized basis from about 5 55 to about 85% by weight of a monomer selected from the group consisting of alkyl acrylate esters and alkyl methacrylate esters containing from 4 to about 12 carbon atoms in the alkyl group and mixtures thereof, from 0 to about 35% by weight of an alkyl acrylate or methacrylate 10 ester containing less than 4 carbon atoms in the alkyl group, from 0.01 to about 2% by weight of a glycidyl monomer, from about 1 to about 10% by weight of an N-vinyl lactam, and from 0 to about 15% by weight of an 15 unsaturated carboxylic acid, said copolymer having a weight average molecular weight of at least about 200,000 and a glass transition temperature less than about -15°C.

2. A pressure-sensitive adhesive as claimed in 20 claim 1 which contains, based on the total weight of monomers, up to about 35% by weight, of a monomer selected from the group consisting of polystyryl ethyl methacrylate, aceto-acetoxy ethyl methacrylate, styrene, alpha olefins, vinyl esters of alkanoic acids containing 25 more than about three carbon atoms and mixtures thereof.

3. A pressure-sensitive adhesive as claimed in 30 claim 1 in which the alkyl acrylate is 2-ethyl hexyl acrylate or isoctyl acrylate.

4. A pressure-sensitive adhesive as claimed in claim 2 in which the alkyl acrylate is 2-ethyl hexyl acrylate or isoctyl acrylate.

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1        5. A pressure-sensitive adhesive as claimed in  
claim 1 in which the glycidyl monomer is selected from  
the group consisting of glycidyl acrylate, glycidyl  
methacrylate, allyl glycidyl ether and mixtures thereof.

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6. A pressure-sensitive adhesive as claimed in  
claim 2 in which the glycidyl monomer is selected from  
the group consisting of glycidyl acrylate, glycidyl  
methacrylate, allyl glycidyl ether and mixtures thereof.

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7. A pressure-sensitive adhesive as claimed in  
claim 3 in which the glycidyl monomer is selected from  
the group consisting of glycidyl acrylate, glycidyl  
methacrylate, allyl glycidyl ether and mixtures thereof.

15

8. A pressure-sensitive adhesive as claimed in  
claim 4 in which the glycidyl monomer is selected from  
the group consisting of glycidyl acrylate, glycidyl  
methacrylate, allyl glycidyl ether and mixtures thereof.

20

9. A pressure-sensitive adhesive as claimed in  
claim 1 in which the N-vinyl lactam is selected from the  
group consisting of N-vinyl pyrrolidone, N-vinyl  
caprolactam and mixtures thereof.

25

10. A pressure-sensitive adhesive as claimed in  
claim 2 in which the N-vinyl lactam is selected from the  
group consisting of N-vinyl pyrrolidone, N-vinyl  
caprolactam and mixtures thereof.

30

11. A pressure-sensitive adhesive as claimed in  
claim 3 in which the N-vinyl lactam is selected from the  
group consisting of N-vinyl pyrrolidone, N-vinyl  
caprolactam and mixtures thereof.

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1 12. A pressure-sensitive adhesive as claimed in  
claim 4 in which the N-vinyl lactam is selected from the  
group consisting of N-vinyl pyrrolidone, N-vinyl  
caprolactam and mixtures thereof.

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13. A pressure-sensitive adhesive as claimed in  
claim 5 in which the N-vinyl lactam is selected from the  
group consisting of N-vinyl pyrrolidone, N-vinyl  
caprolactam and mixtures thereof.

10

14. A pressure-sensitive adhesive as claimed in  
claim 6 in which the N-vinyl lactam is selected from the  
group consisting of N-vinyl pyrrolidone, N-vinyl  
caprolactam and mixtures thereof.

15

15. A pressure-sensitive adhesive as claimed in  
claim 7 in which the N-vinyl lactam is selected from the  
group consisting of N-vinyl pyrrolidone, N-vinyl  
caprolactam and mixtures thereof.

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16. A pressure-sensitive adhesive as claimed in  
claim 8 in which the N-vinyl lactam is selected from the  
group consisting of N-vinyl pyrrolidone, N-vinyl  
caprolactam and mixtures thereof.

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17. A pressure-sensitive adhesive as claimed in  
claim 1 in which the formed copolymer is cross-linked by  
exposure to heat, ionic additive, actinic radiation or  
electron beam radiation.

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1 18. A pressure-sensitive adhesive comprising a  
copolymer comprising from about 55 to about 85% of an  
alkyl acrylate ester selected from the group consisting  
of isooctyl acrylate and 2-ethyl hexyl acrylate, from  
5 about 0.01 to about 2% by weight glycidyl methacrylate,  
from about 1 to about 10% of a N-vinyl lactam selected  
from the group consisting of N-vinyl pyrrolidone and N-  
vinyl caprolactam, from about 5 to 13% by weight  
unsaturated carboxylic acid, and up to about 35% by  
10 weight methyl acrylate, said polymer having weight  
average molecular weight from about 200,000 to about  
500,000 and a glass transition temperature less than  
about -15°C.

15 19. A pressure-sensitive adhesive as claimed in  
claim 18 in which the formed copolymer is cross-linked  
by exposure to heat, ionic additive, actinic radiation  
or electron beam radiation.

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Fig. 1

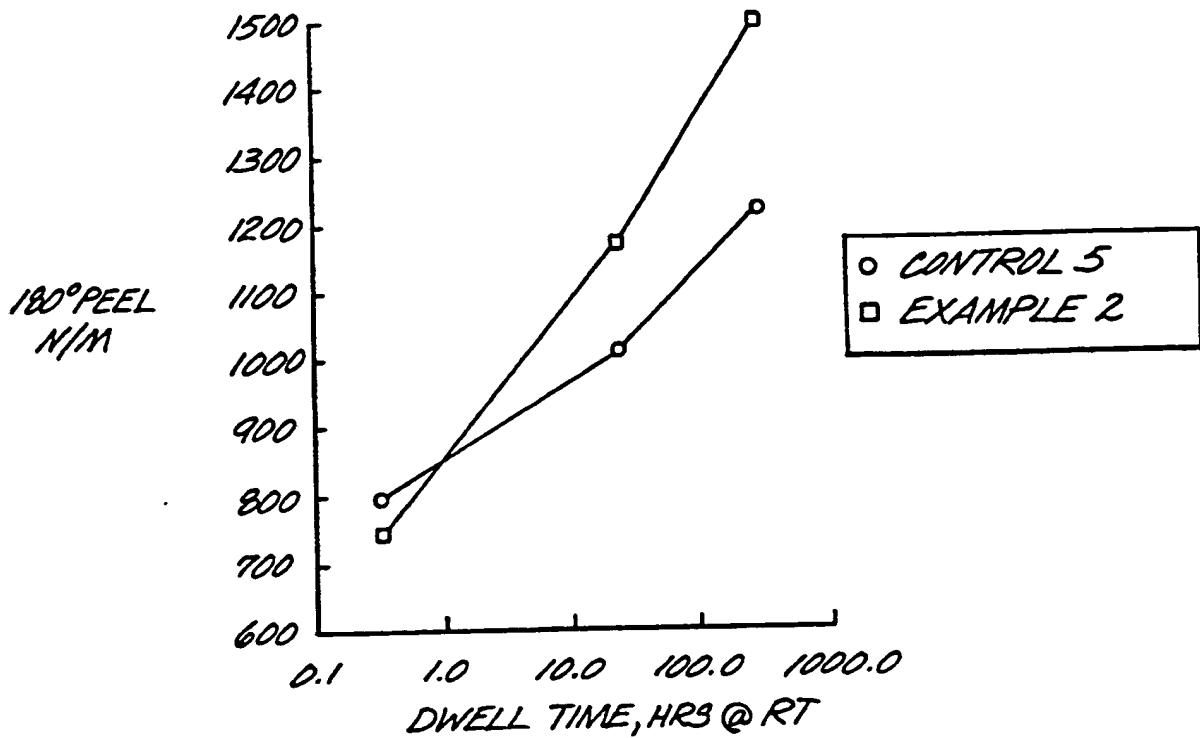
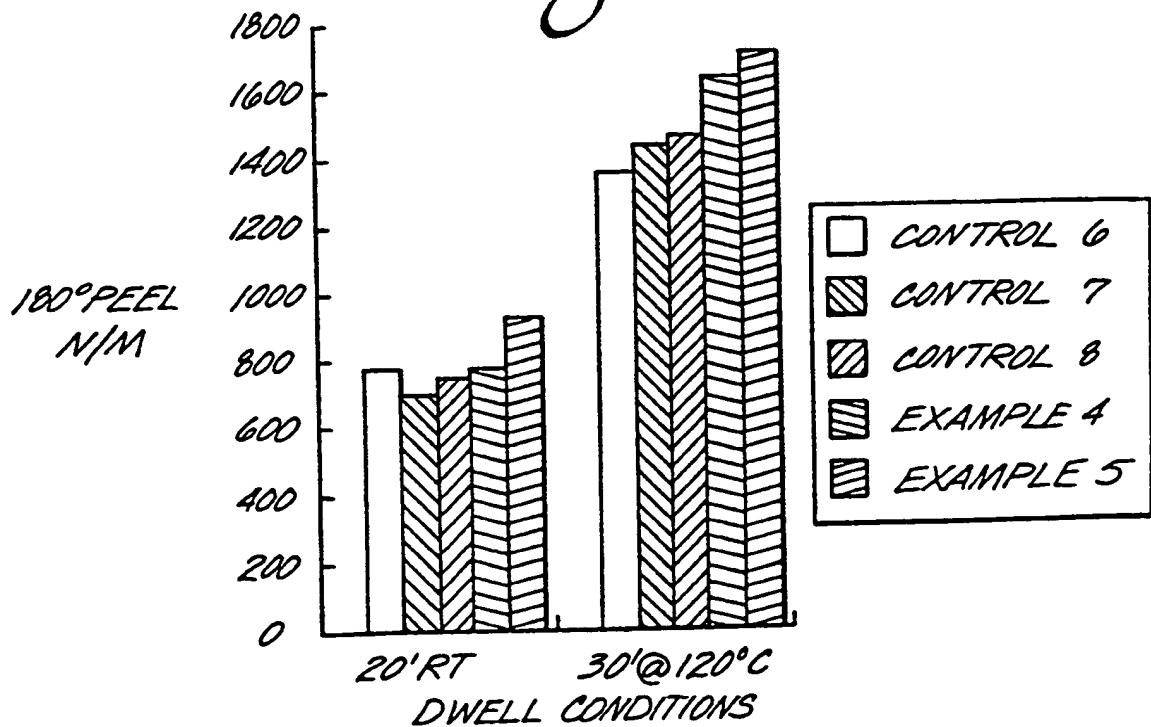


Fig. 2



# INTERNATIONAL SEARCH REPORT

International Application No. PCT/US88/04588

## I. CLASSIFICATION OF SUBJECT MATTER (if several classification symbols apply, indicate all) <sup>6</sup>

According to International Patent Classification (IPC) or to both National Classification and IPC  
 Int Cl <sup>4</sup> CO8F, 26/10; 24/00; USCl: 526/264,273

## II. FIELDS SEARCHED

Minimum Documentation Searched <sup>7</sup>

Classification System	Classification Symbols
USCl	526/264,273

Documentation Searched other than Minimum Documentation  
 to the Extent that such Documents are Included in the Fields Searched <sup>8</sup>

## III. DOCUMENTS CONSIDERED TO BE RELEVANT <sup>9</sup>

Category <sup>10</sup>	Citation of Document, <sup>11</sup> with indication, where appropriate, of the relevant passages <sup>12</sup>	Relevant to Claim No. <sup>13</sup>
A	US, A 3,787,380, STAMBERGER 22 January 1974; See Entire Document	1-19
A	US, A 4,510,197, SHAH 09 April 1985; See Entire Document	1-19

\* Special categories of cited documents: <sup>10</sup>

"A" document defining the general state of the art which is not considered to be of particular relevance

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"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step

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"&" document member of the same patent family

## IV. CERTIFICATION

Date of the Actual Completion of the International Search

24 February 1989

Date of Mailing of this International Search Report

18 APR 1989

International Searching Authority

ISA/US

Signature of Authorized Officer

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